

ELECTRETS FORMED FROM POLYETHYLENE TEREPHTHALATE FILMS

Dr. L. M. Baxt and Dr. L. D. Grandine of the Philip Morris Research Center disclosed to me in July 1962, the results of their research on the use of polyethylene terephthalate films to prepare electrets.

The term "electret" was coined by Oliver Heaviside to denote a permanently electrified substance exhibiting electrical charges of opposite sign on opposite faces. The electrification is through the volume of the substance in contrast to surface electrification and hence is a volume effect.

The first man to prepare an electret was Eguchi¹ who melted equal parts of carnauba wax and resin, with the addition of some beeswax, and permitted the substance to solidify in a strong electric field. Since then, many papers have been published on the subject and a few patents have been obtained on methods of preparing electrets from specific substances (for example, Padgett, U. S. 2,986,524; Gemant, British 435,950, 438,672; German 518,087; Nicol, U. S. 2,612,966; Minto, U. S. 3,028,864.)

In general the substances which have been deemed capable of permanent electrification are those which remain insulators when heated, and which have moderate or high dielectric absorption. These materials are polar dielectrics as opposed to

¹M. Eguchi, "On the Permanent Electret," Phil. Mag. 49, 178 (1925).

non-polar or slightly polar. Nearly all workers thus far have used waxes, resins, or polymers.

Drs. Birt and Grondine have now discovered that films of the polymer polyethylene terephthalate can be made into electrets by methods of electrification somewhat similar to those used by Iguchi and others but that the electrets thus formed have unique properties, properties not possessed or not possessed to the same degree by other known electrets.

Charges on electrets are measured in coulombs/square centimeter. The charges reported for electrets of the waxes and resins (for example, carnauba wax and beeswax, nylon, polymethyl methacrylate, polytetrafluoroethylene electrets,) and for some common ceramics and the like are at most 4 or 5×10^{-9} . In the polyethylene terephthalate electrets of this invention, charges more than triple that order (16×10^{-9}) have been measured.

According to present day theory, electrets acquire two sets of opposing electric charges. One of these charges is temporary and is called the heterocharge. The other charge, known as the homocharge is relatively permanent. Therefore, in normal electret behavior (as the behavior of electrets of carnauba wax) the charge which decays is the heterocharge; the more permanent charge is the homocharge. On the basis of our results thus far, the opposite seems to be true of the electrets of polyethylene terephthalate; the homocharge decays and the heterocharge apparently is the more permanent charge.

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Another phenomenon observed with electrets of polyethylene terephthalate has to do with the measurement of its drop-off of charging current. The drop-off with other electrets has followed a pattern of a continuous current decrease. Sometimes the polyethylene terephthalate electret exhibits a sharp rise in charging current and a subsequent slow decrease.

For example, when an electret-forming material (e.g., carnauba wax, acrylics, etc.) is placed between two electrodes and a strong electric field is applied (approximately 8-11 kilovolts per centimeter), normally a forming current pattern like this has been reported:

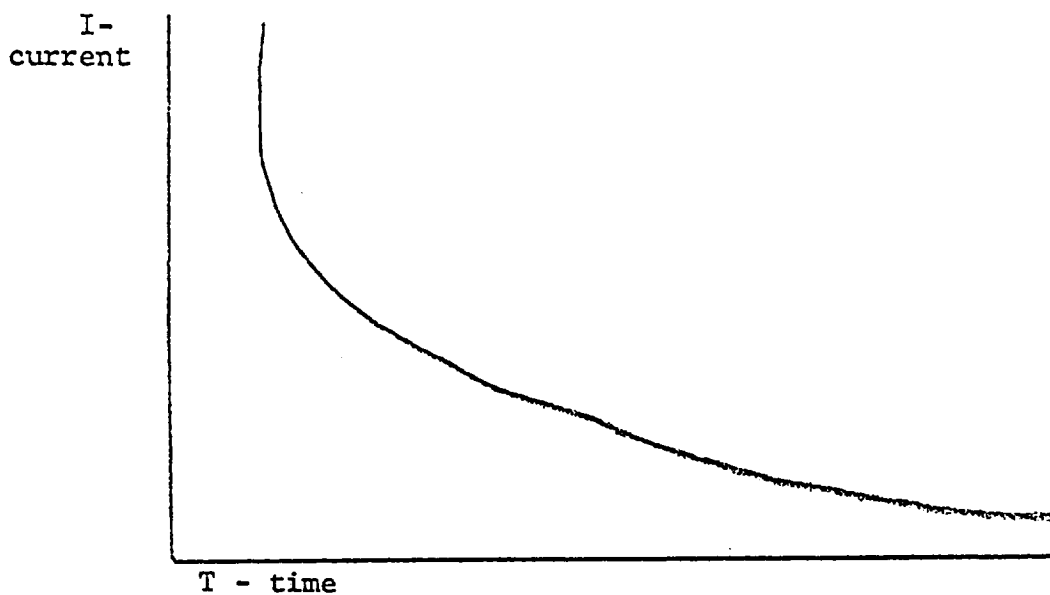


Figure 1

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With the electrification of polyethylene terephthalate, at field strengths of 120-200 kilovolts per centimeter, the forming pattern differs in this way:

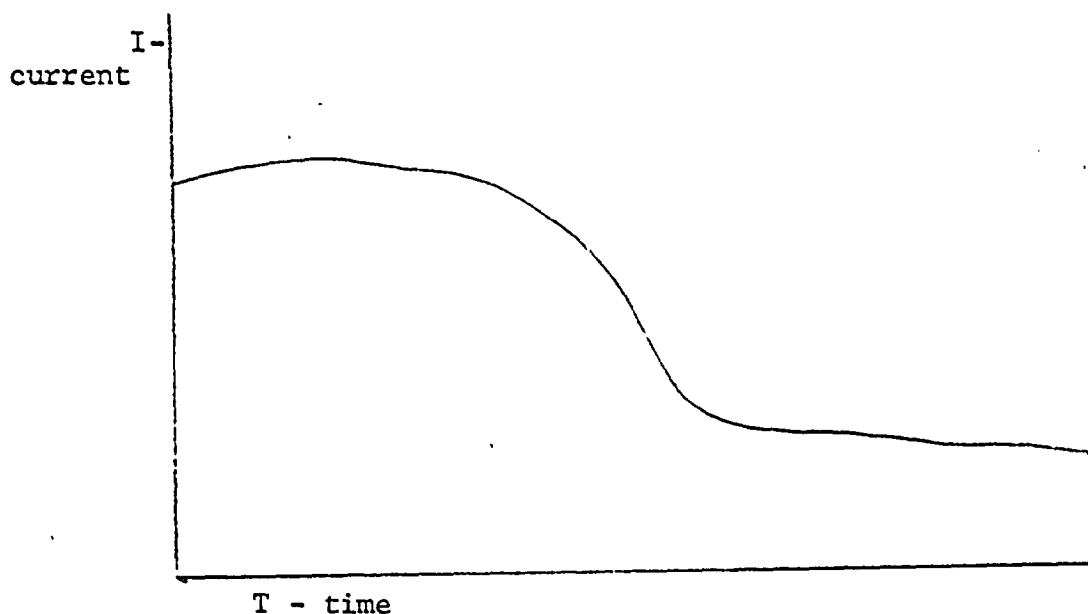


Figure 2

In conventional methods of preparing electrets, a molten dielectric is allowed to solidify between two electrodes across which a high potential is applied. It is not essential that the material be melted. It can be heated and then cooled under an electric field.

The heterocharge in a freshly made electret exceeds the homocharge; charge measurements give the difference between these two. The heterocharge or normally temporary charge is positive on the side of the electret which was in contact with the negative electrode and negative on the side which was in contact with the positive electrode: hence the name "heterocharge," which means "different charge;" that is, different from the charge of the forming electrode.



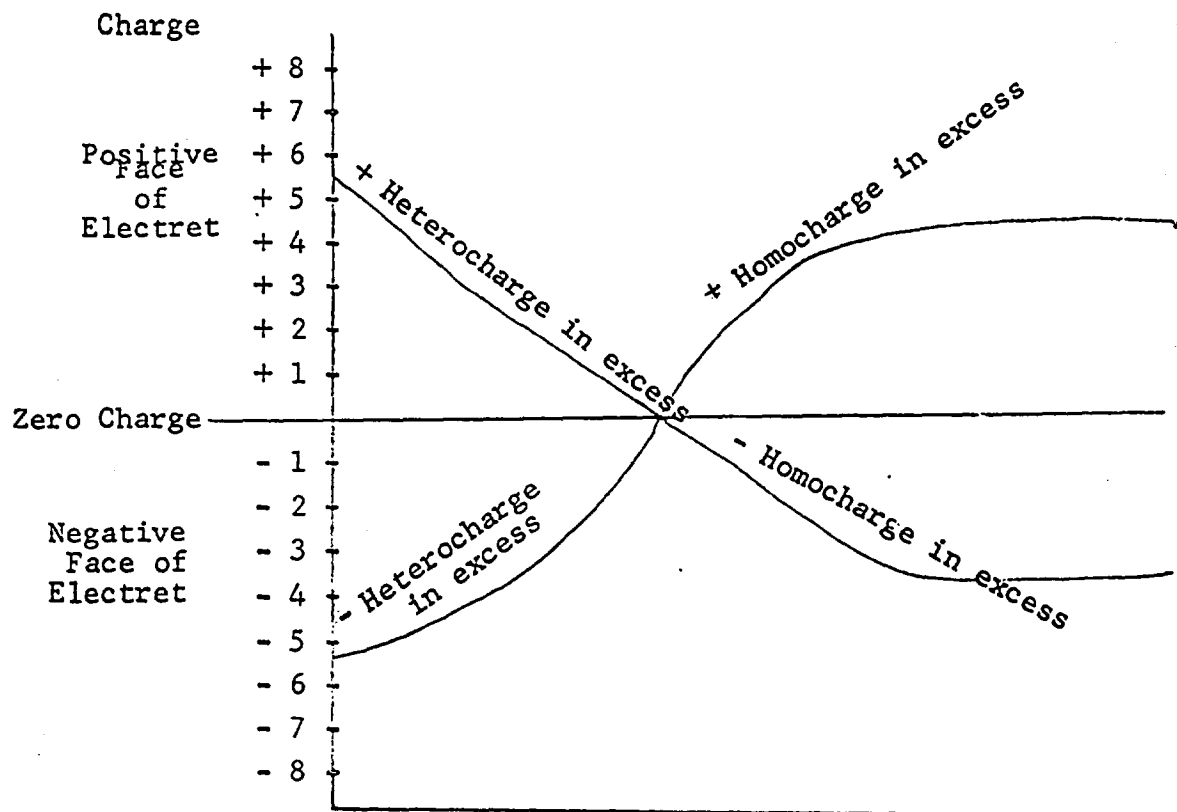
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The homocharge is positive on the side of the electret which was in contact with the positive electrode during manufacture and negative on the side which was next to the negative electrode; hence the name, "homocharge" which means "same charge." The homocharge is thought to be caused by sprays of electrical charges which leave the forming electrodes and enter the electret material because of the high potential which is present. All of these forms of polarization are "frozen in" when the electret material hardens.

Drs. Baxt and Grandine have now discovered that, by the use of extremely high fields applied to polyethylene terephthalate film, an electret can be formed which does not have a reversal of charges. The ordinary electret cannot be put into use until after the charge reversal has taken place. The polyethylene terephthalate electret of this invention can be put into use immediately, since the electret does not go through a point of zero net charge. Instead of following normal behavior, the initial heterocharge of the polyethylene terephthalate electret continues to increase and shows no charge reversal over extended storage periods. Diagrams of normal electret behavior and the behavior of the polyethylene terephthalate electret are shown below:

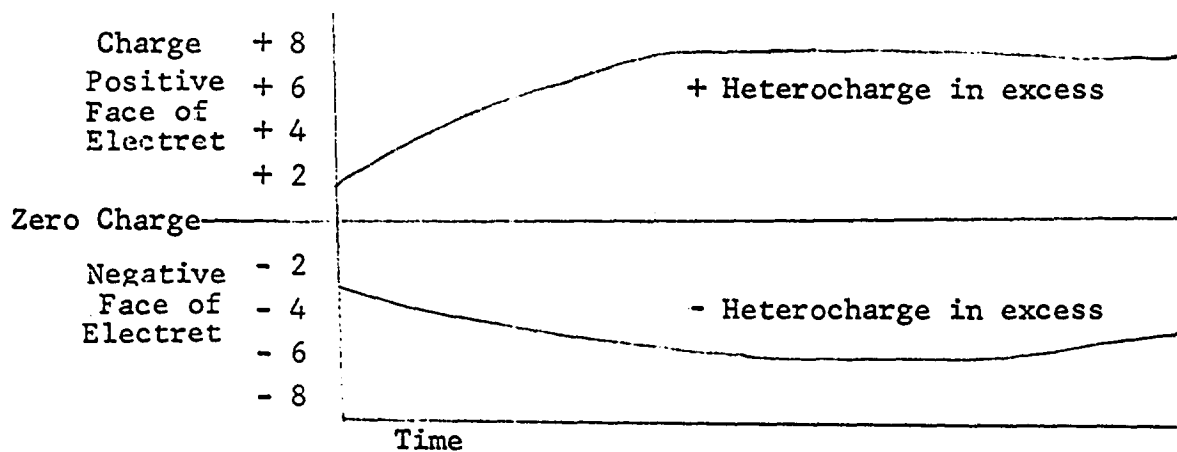
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Normal Electret Behavior

Figure 3



Behavior of Electret of Polyethylene Terephthalate

Figure 4

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Although polymer electrets have been formed previously, they have had a life of only a few months. This polymer electret (polyethylene terephthalate) has a rate of decay that makes it effective almost indefinitely.

The method of forming the electret of this invention differs from the early methods in that the electret material is treated at a temperature 30 to 40° below its melting point instead of being charged while in a molten state. Heretofore voltages on the order of 8 - 11 kilovolts per centimeter were used in forming electrets. For this invention voltages of 120-200 kilovolts per centimeter were used.

The inventors have also discovered a method of stacking the electret-forming material so that a number of disks can be formed at one time. When a package of four disks is placed in the forming field for long periods (3 - 7 days), the disk adjacent to the positive electrode has a much lower charge than the other three disks. The three disks have charges on the order of 10 to 14 x 10⁻⁹ coulombs/sq. cm. For shorter times of electrification (2 - 30 hours), the charge is practically equal for all the disks in the package.

The polyethylene terephthalate electret, because of its unique properties, has utility beyond that of electrets now known. It can be used in any device where a permanent electrostatic charge is required and it can be put to use immediately upon production without the delay necessary with other electrets. It can be used in electrostatic filters, dosimeters, microphones, electrometers, vibration meters, and similar devices. It is particularly applicable where such devices of low weight are needed

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(for example, in space research).

The invention of Drs. Baxt and Grandine can be illustrated by the following examples:

Example 1

A polyethylene terephthalate disk four inches in diameter and 10 mils thick was placed between aluminum foil disks and the combination was placed between heavy stainless steel disks. The stainless steel was to serve as electrodes for the conduction of high voltage current. This assembly was placed in an oven equipped with temperature controls, with a fan for equalization of internal temperature, and with a high voltage direct current power supply. The oven during the electret formation was operated under atmospheric pressure and at a temperature of 150°C. The faces of the dielectric disk were shorted during heating, before application of field, to remove any residual charges present.

The bottom aluminum electrode was connected to the negative voltage output of the power supply and the top electrode was connected through an electrometer to ground. When the sample temperature reached 150°C., that temperature was maintained for one hour. Then the voltage was applied (a field strength of 197 KV/cm) and the current was measured periodically during the forming period of 20 hours. The measurements of current given below show a sharp rise and a subsequent gradual decrease in the forming current, which seems to be unique in the field of dielectrics. Ordinarily, only decreases are observed.

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TABLE I

Temperature was held constant at $150^{\circ} \pm 0.5$

<u>Time</u>	<u>Charging Time (Minutes)</u>	<u>Current (μa)</u>
1:27	0	0
1:42	15	4.8
1:44	17	5.0
2:05	78	5.7
2:40	113	5.1
3:12	185	4.8
3:33	206	4.7
4:05	266	4.5
4:40	313	4.4
8:35	1148	1.3
9:28	1201	1.3

At the end of the 20-hour charging period, the heater in the oven was disconnected, the door opened, and the disk was allowed to cool while still in the electric field. The electric field was removed and the disk was removed from the oven, and the surface charge on the disk was measured by placing it in a dissectible capacitor; i.e., with one movable electrode. The electrode was placed in contact with the electret, the system was grounded to remove any charge, and the electrode was moved away from the electret to a predetermined distance. The resulting induced charge on this electrode was measured by the use of an appropriate capacitor and electrometer. The voltage reading of the electrometer was then converted to charge density by the

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use of the formula given below:

$$Q = \frac{CV}{A}$$

where Q is charge density

C is capacitance of the capacitor used

A is area of the movable electrode

V is voltage read on the electrometer

For this particular electret the voltage reading for the top of the disk was +3.35 volts. For the bottom, the reading was -2.36 volts. The area of the movable electrode was 12.56 cm^2 . The capacitance of the capacitor was $.01 \times 10^{-6}$.

$$\text{Top} \quad - \quad Q = \frac{(.01 \times 10^{-6}) \ 3.35}{12.56} = -2.7 \times 10^{-9} \text{ coulombs/cm}^2$$

$$\text{Bottom} \quad - \quad Q = \frac{(.01 \times 10^{-6}) \ 2.36}{12.56} = +1.9 \times 10^{-9} \text{ coulombs/cm}^2$$

This calculation shows that the freshly-made electret had a heterocharge in excess. The electret was kept shorted and desiccated in storage. The data given below show that over about a three month storage period, the heterocharge in excess of the polyethylene terephthalate electret increased, a phenomenon heretofore not exhibited by known electrets.

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TABLE II

<u>Elapsed Time (hours)</u>	<u>Positive Electrode Heterocharge Top</u>	<u>Negative Electrode Heterocharge Bottom</u>
0	-2.7	+1.9
3	-2.8	+3.8
6	-2.9	+3.6
28	-5.1	+3.2
49	-3.9	+4.0
173	-6.0	+5.5
292	-5.4	+4.0
547	-7.0	+3.9
2163	-8.4	+8.7
9818	-8.0	+7.9

Electrets have been used in many applications - as electrostatic filters, microphones, electrometers, etc. Perhaps one of the best applications is in a dosimeter. This application was carried out in the manner described below.

A polyethylene terephthalate electret similar to the one described above was subjected to a polonium source (α radiation) of 0.5 millicurie strength. First, the electret was placed for slightly over one hour in intimate contact with the surface of the source and its charge measured. Then the electret was separated from the radiation source by suspending the source three millimeters above its surface. A non-irradiated control electret was measured for charge each time the test electret was measured. The attached graph and Table III show the data obtained from this experiment.

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TABLE III

Direct Contact

Time Treated Min.	Total Time Treated Min.	Charge 10^{-9} Coul./cm ²			
		<u>Irradiated Sample</u>		<u>Control Sample</u>	
		Top Value	Bottom Value	Top Value	Bottom Value
0	0	-10.24	+10.24	-10.24	+10.24
110	110	-9.46	+9.46	-10.24	+10.24
3950	3950	-6.31	+6.31	-10.17	+10.17

3 MM Separation

0	3950	-6.31	+6.31	-10.17	+10.17
1440	5390	-4.65	+4.57	-10.30	+10.40
2550	6500	-4.54	+3.54	-10.17	+10.17

As shown above, the polyethylene terephthalate electret can be used as a dosimeter, a simple device for detecting radiation. It illustrates well the sensitivity of electrets toward radiation. These electret dosimeters have an advantage over other dosimeters now in use in that they eliminate the necessity for elaborate encasing materials such as must be used with other types of dosimeters. For example, dosimeters in which photographic film is used must have light-proof casings. The electret dosimeter requires no encasement.

Also the discharge of the electret can be readily detected simply by passing it into a dissectible capacitor. With the photographic-type dosimeter, time is required for the film to be developed.

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Example 2

A polyethylene terephthalate disk four inches in diameter and 10 mils thick was shorted and grounded then treated as described in Example 1. Voltage was applied (a field strength of 197 KV/cm) and the current was measured periodically during the forming period of 90 hours. After removal from the oven the measurements of charge at different times were as follows:

<u>Elapsed Time</u> (Hours)	<u>Surface Charge Density</u> 10^{-9} coul/cm ²	
	<u>Top</u>	<u>Bottom</u>
0	-8.0	+ 8.0
1	-7.8	+ 8.1
26	-7.9	+ 7.2
120	-8.0	+ 8.4
240	-9.1	+ 8.0
312	-9.6	+ 8.8
984	-11.5	+10.8
2700	-11.5	+ 9.0
5600	-11.5	+10.4

This electret, charged for a longer period of time than the one described in Example 1, had a much higher initial charge and increased only slightly in charge over a period of nearly 6000 hours. This example, combined with the electret of Example 1, illustrates the importance of proper selection of charging time and voltage upon the constancy of charge of the electret. This

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is readily observed in the figure below:

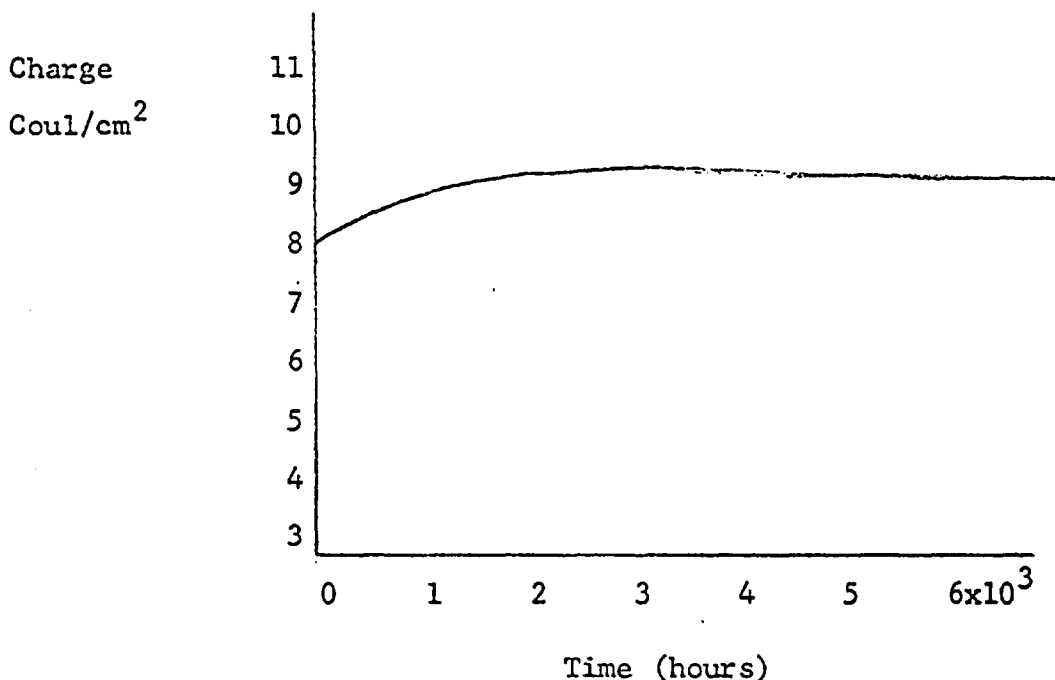


Figure 5

Example 3

Four polyethylene terephthalate disks four inches in diameter and 10 mils thick were shorted individually and grounded to put both surfaces at ground potential. This package of disks was then placed between aluminum foil disks and the combination was treated as was the electret formed in Example 1, except that a field strength of 49 KV/cm was used and the disks were changed for two hours. The net charge density of the four disks is shown below:

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	<u>-15- 10^{-9} coul/cm²</u>	
	<u>Top Face</u>	<u>Bottom Face</u>
Top Disc # 1	-7.3	+8.4
Disc #2	-7.8	+8.3
Disc #3	-7.9	+8.7
Bottom Disc #4	-8.6	+7.1

Figure 6

The charge density of the four disks show uniformly high heterocharge throughout the stack. This illustrates that by proper selection of charging time at a given field strength uniformly high heterocharges can be obtained in a stacked package of disks.

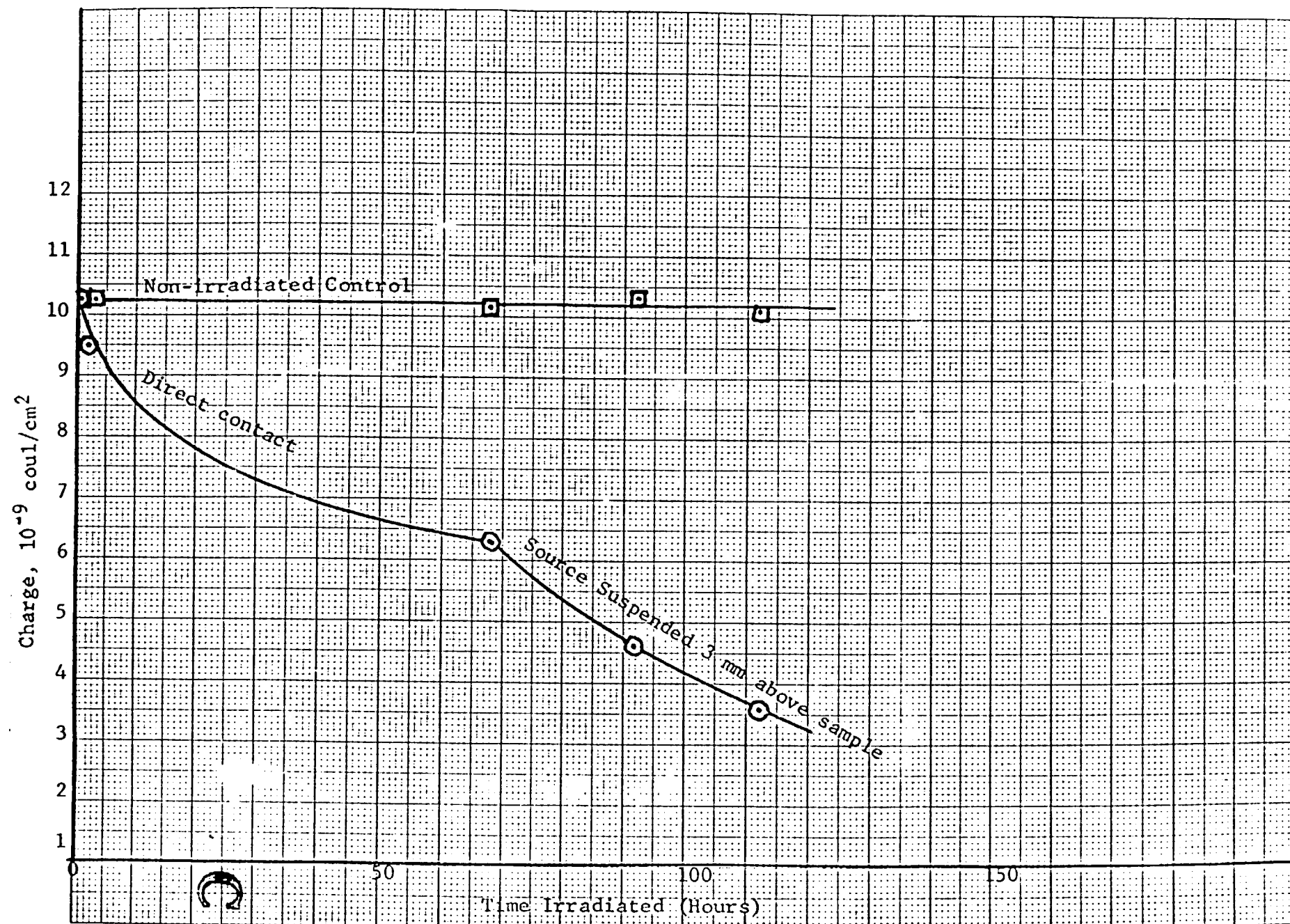
The results of any further development of the investigation of electrets formed from polyethylene terephthalate films will be reported and shall become a part of this disclosure.

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